## **Supporting Information**

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## SI Text

Cutting Procedure for SWNT Electrode Formation. After device fabrication and characterization, a PMMA layer (950, A2) was spincast (4000 PRM, 45 s) on the surface and then baked at 170 °C for 2 min. Using e-beam lithography, we run a DesignCAD file with  $\approx$ 5-nm-width line at the specific position to obtain the window precursor. Then the mixture of water/ isopropanol (1:3) is used for the liftoff at 5 °C for 1 min with the aid of sonication. After liftoff, the devices were washed by DI water and dried with a stream of N2 gas. After the window was opened, the devices were put into a TECNIQUES Series 800 RIE machine. The nanotubes were then locally cut through the open window by oxygen plasma (50 W RF power, oxygen 250 mTorr, for 10 s). After cutting, the devices were soaked in acetone solution overnight, removed, washed by acetone, isopropanol, and deionized water and dried with a stream of N<sub>2</sub> gas. Under these optimized conditions,  $\approx 20-25\%$  of the tubes were completely cut. Based on the previous results, the statistical variability of the plasma etch process creates ensembles of nanotube devices with gaps in the 1- to 10-nm range. Parts of the devices were used further to fabricate the devices with SWNT-Metal junctions protected by HSQ using e-beam lithography according to our previous report.

 $\label{eq:current-Voltage Curves for a Device Made by Drop-Casting. See Fig. S1.$ 

Device Characteristics for a Metallic Device. See Fig. S2.

Power-Dependence Experiment. See Fig. S3.

**Wavelength-Dependence Experiment.** To further understand the important role of HBCs in device photoconductivity, we intend to display the wavelength dependence measurement of the devices. In the source-drain current ( $I_{sd}$ ) vs. wavelength experiments, the device was illuminated with visible light (370–700 nm) coming from a xenon lamp (Oriel Instruments). The light

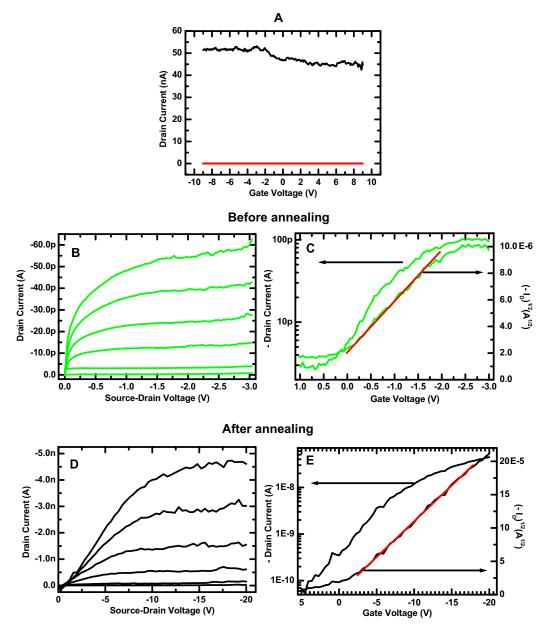
was collimated and focused with mirrors and lens from Newport Corporation and diffracted with monochromator of ISA Instruments. The outcoming light was filtered with a long-pass filter (>365 nm) to eliminate double-light phenomena. The light power was measured with 1830C Newport power meter. Source and drain potentials were applied to the transistor with 2 source meters (2400; Keithley), functioning as a semiconductor parameters analyzer. The photocurrent was measured by the Keithley source meters. The various instruments (monochromator, source meters) were controlled by a home-made Labview program (National Instruments) as well as the data collection using a PC.

The current responses as a function of light wavelength of a device made by drop-casting have been shown in Fig. S4 while the device is held at -20-V source-drain bias and 0-V gate bias. The red curve shows the results of the wavelength-dependence experiments. The black curve indicates the UV/vis absorption spectrum of HBC thin films on quartz. The peaks of the photocurrent spectral response of the device at  $\approx 400$  nm match those of the UV/vis absorption spectrum of HBC thin films. Because of the combinations of vibronic excitations, there are several overlapping peaks. The weak peaks at >450 nm in the red curve are most likely due to (radialene  $\pi$ ) – (radialene  $\pi^*$ ) triplets. These results clearly prove that self-assembled columnar nanostructures of HBC molecules play the key role in device characteristics.

Current–Voltage Curves for a Device Made from 6,13-di(2'-thie-nyl)pentacene. See Fig. S5.

Current-Voltage Curves for Another Device Made from 6,13-di(2'-thienyl)pentacene. See Fig. S6.

Photoresponsibility for the Same Device as in Fig. S6. See Fig. S7.



**Fig. S1.** Device characteristics of a device made by drop-casting before and after annealing. (A) Electrical characteristics of a metallic tubes ( $I_D$  vs.  $V_G$  at  $V_D$  = 50 mV) used for testing before (black curve) and after (red curve) oxidative cutting. (B) Transistor output,  $V_G = 0$  to -3 V in 0.6-V steps. (C) Transfer characteristics for the device,  $V_D = -3$  V. (D) Transistor output,  $V_G = 0$  to -20 V in 4-V steps for the same device after heat treatment. (E) Transfer characteristics for the device,  $V_D = -20$  V.

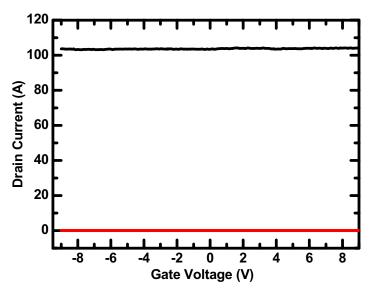
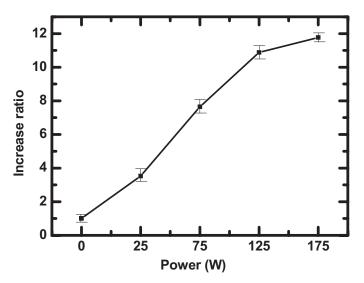


Fig. S2. Electrical characteristics of a metallic tube (I<sub>D</sub> vs. V<sub>G</sub> at V<sub>D</sub> = 50 mV) used for testing in Fig. 4 before (black curve) and after (red curve) oxidative cutting.

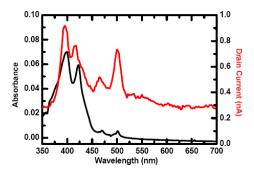
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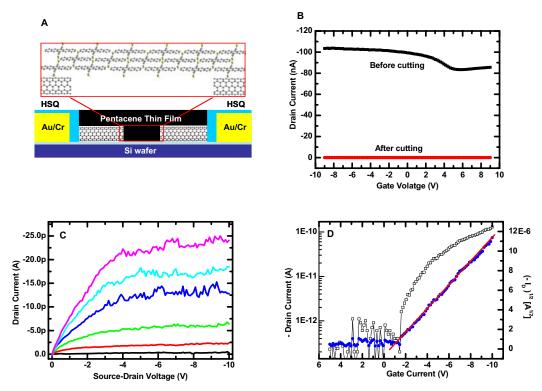


**Fig. S3.** The power dependence of the photocurrent of a device when the power of the halogen lamb was gradually increased to its maximum while the source-drain and gate voltage bias was fixed at -20 V. With the increase of light power, the drain current of the device gradually saturates, indicating that the photo-induced carrier density reaches its maximum.

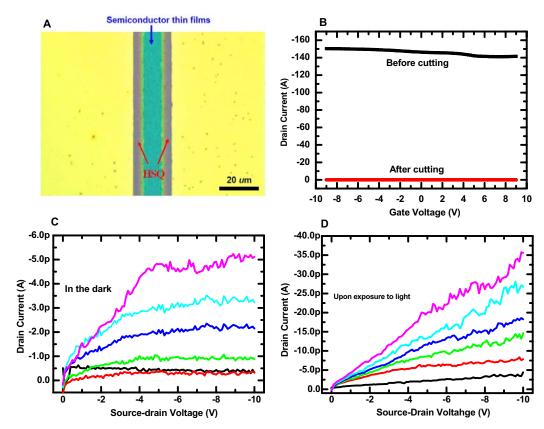
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**Fig. S4.** Demonstration of the wavelength dependence of a device made by drop-casting. The red curve shows the wavelength dependence of the current responses of the device while the device is held at -20-V source-drain bias and 0-V gate bias. The black curve shows the UV/vis absorption spectrum of HBC thin film on quartz.



**Fig. 55.** All of the working devices exhibit the typical *p*-type FET properties. (*A*) The schematic of the device structure used for testing 6,13-di(2'-thienyl)pentacene with SWNT-molecule junctions protected by HSQ. (*B*) Electrical characteristics of a metallic tube ( $I_D$  vs.  $V_G$  at  $V_D$  = 50 mV) used before (black curve) and after (red curve) oxidative cutting. (*C*) Transistor output of the same device,  $V_G$  = 0 to -10 V in 2-V steps. (*D*) Transfer characteristics for the device,  $V_D$  = -10 V.



**Fig. S6.** Device characteristics of another device made from 6,13-di(2'-thienyl)pentacene. (*A*) An optical photograph of a device used for testing 6,13-di(2'-thienyl)pentacene with SWNT-metal junctions protected by HSQ. (*B*) Electrical characteristics of a metallic tube ( $l_D$  vs.  $V_G$  at  $V_D$  = 50 mV) used before (black curve) and after (red curve) oxidative cutting. (*C*) Transistor output,  $V_G$  = 0 to -10 V in 2-V steps in the dark. (*D*) Transistor output,  $V_G$  = 0 to -10 V in 2-V steps under light irradiation.

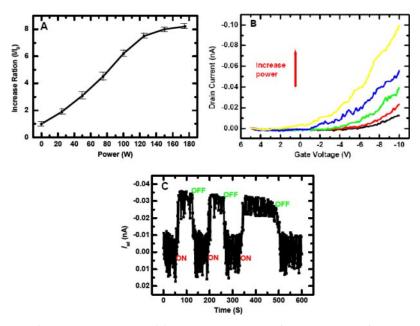


Fig. 57. Photoresponsive behaviors of the same device in Fig. S6. (A) The power dependence of the photocurrent of the same device measured in Fig. S6 when the power of the halogen lamb was gradually increased to its maximum while the source-drain and gate voltage bias were fixed at -10 V. (B) Transfer characteristics of the same device with the increase of light power. (C) The drain current as a function of time while the same device is held at -10-V source-drain bias and -10-V gate bias by switching on/off light.